

# DOMINION OF CANADA

THE HONORARY ADVISORY COUNCIL FOR SCIENTIFIC AND  
INDUSTRIAL RESEARCH

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REPORT No. 3

## THE DE-TARRING OF GAS BY ELECTRICAL PRECIPITATION

BY

J. G. DAVIDSON, Ph.D.



CANADA

Published by the authority of the Sub-Committee of the Privy Council  
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## THE DE-TARRING OF GAS BY ELECTRICAL PRECIPITATION.

The work described herein was done at various times during the last four years, in intervals of university duties. Its scope was limited by financial and other exigencies.

In the Northwest, where work was begun, there exist only a few small distillation plants. The meagre facilities afforded by these plants rendered an exhaustive investigation impracticable. Fortunately, however, with the financial and technical assistance of the Honorary Advisory Council for Scientific and Industrial Research, it became possible last winter to continue and complete the work at the by-product coke-oven plant of the Algoma Steel Corporation at Sault Ste. Marie, Ont. In thus rounding out the series of experiments the problem was brought to a stage where the attention of those interested in the matter industrially could be at once engaged.

This paper, issued under the auspices of the Honorary Advisory Council, constitutes the first published report of any portion of the investigation.

### THEORY OF ELECTRICAL PRECIPITATION.

Electrical precipitation is the process of removing suspended particles of smoke, or fume, or "fog" from a gas, by electrifying the particles static, and then separating them from the gas by attraction to a surface charged with an opposite potential.

As is well known, a charge of electricity has greater intensity at the edges or at extruding points of a conductor than on the smooth surfaces. As the potential rises through repeated charges of electricity, a point is reached where there occurs a discharge into the gas. This takes place from the edges or points above mentioned, rather than from the smoother surfaces. The discharging surface is covered with a velvety glow when negative electricity is being discharged; whilst the discharge of positive electricity manifests itself as a brush reaching out from the surface. Particles of the substance suspended are electrified by this discharge, the gas itself not becoming charged.

Electrical precipitation installations consist of one or two pairs of electrodes, between which the gas, with its suspended particles, is made

to pass. The smaller, or discharge electrodes, may or may not have sharp points or edges to facilitate the discharge. The larger, or collecting, electrode is a flattened surface from which there is as small an electrical discharge as possible.

The installations, or treaters, that have been developed commercially are of two principal types, the pipe treater and the plate treater, the general action of both types being similar.

In operation, insulated wires are suspended down the centres of the pipes in the pipe treater and between the plates in the plate treater. The gas to be treated is so distributed to the pipes or plates as to ensure the removal of all suspended matter during its passage through each unit. For reasons that are more or less obscure, and which need not be discussed here, the discharge electrodes are in practice charged negatively to a potential which is maintained at a point just below that at which sparks would pass from the wire to the plate. The size of the pipe or the distance between the plates will thus depend upon the potential that may be conveniently maintained upon the discharging wires.

The most common diameter of pipe, or distance between plates, is about 12 inches. The potential that must be maintained for such distances varies with the chemical composition of the gas and with the temperature, from, say 50,000 to 100,000 volts. When the treater is working properly the current consumption consists of the silent or glow discharge from the discharging electrode (which is the only power that is usefully employed) together with the charging currents of the electrical machinery and the leakage over the insulators.

As the gas passes through the pipes or between the plates, suspended particles, either liquid or solid, are electrified by the discharge from the wires. The negatively electrified particles are then repelled from the negative wire and attracted to the surface of the plate or pipe that is charged positively.

It may be mentioned here that the discharge from the wire is accompanied by an electrical windage which assists to some extent in driving the suspended particles towards the collecting surface. If the suspended matter consists of particles of solid material, it will be collected as an adhering dust, and may be shaken off the pipe or plate by rapping. If the suspended material is in the form of liquid particles of fog or mist, it will, of course, run down the pipe or plate and out of the bottom of the treater.

#### DEVELOPMENT OF ELECTRICAL PRECIPITATION.

For nearly a century the principles involved in the electrical precipitation of finely divided liquids and solids from gas have been known to science. The successful commercial application of these principles has depended upon the development of modern electrical machinery.

Attempts were made to develop a commercial process in the "eighties"; but in those days the friction, or static, machine was the only means of obtaining potentials high enough to give electrical discharges from points or small surfaces. The success of the process in recent years has been made possible by the development of the modern high potential transformer, and by the invention of a rectifier to change the alternating current from the transformer into direct current, so that the discharge from the wires might be kept either positive or negative as desired.

The invention of the rectifier and the development of the various other features of the process are to be credited to Dr. F. G. Cottrell, and to numerous later investigators. In recognition of Dr. Cottrell's actual reduction of the theory of electrical precipitation to commercial practice, the method is commonly called the Cottrell Process.

The process was first used in the separation of sulphuric acid mist from the vapours evolved from the parting kettles in the metal refinery of the Selby smelter on San Francisco bay. Efforts were then directed toward collecting the valuable flue-dust from the gases passing into the smelter stacks; but the earlier installations for this purpose, namely, those at the Selby and Balaklava smelters in California, were not wholly successful. Most of the later installations handling smelter gases, however, have proved exceedingly efficient.

The process was first successfully employed on a large scale in the collection of dust carried in the gases from the kilns of the Riverside Portland Cement Company. Here for the last eight years there has been collected not less than 95 per cent of the dust carried in approximately a million cubic feet of gas per minute. For a considerable time the dust thus collected amounted to the enormous total of about one hundred tons daily. Later improvements in handling the kilns has materially reduced this amount.

It should be mentioned in passing that it was soon discovered that a substantial proportion of potash-bearing material in the raw mix was volatilized in the kilns and collected as a part of the dust in the

treaters. This has meant the development of an unexpected source of potash in America. Many cement companies are now installing the process, not as a means of eliminating the dust nuisance, but to effect the recovery of the potash content of the dust.

Of the many hundreds of electrical precipitation treaters installed in many different kinds of industrial plants, practically all are operated at the end of the manufacturing process, where gases generated in the plant were being allowed to escape and carry with them solid or liquid particles in suspension. In many cases the real object has been to avoid litigation by removing the smoke nuisance; though in practically every instance it has been found that the collected material is valuable enough to warrant the installation as a commercial proposition in itself.

During the last three or four years developments have been in the direction of including electrical precipitation as an integral part of various manufacturing processes, and not as a supplementary addition. The collection of cement kiln dust is being modified to meet the market demand for a material containing as great a proportion as possible of soluble potash. It would appear that processes of manufacturing such products may become a separate industry; whereas now they may be said to be incidental to the production of cement.

A great deal of successful experimental work has been done in the smelting of metallic ores by volatilizing the metallic contents and collecting by means of electrical precipitation. This method has been brought to a point where it is being undertaken commercially.

Three great industries, the cement, the sulphuric acid, and the smelting industries give evidence of the waxing importance of electrical precipitation. The investigations described in this bulletin were undertaken with the belief that this method of precipitation may well become the central feature of new collection processes in a fourth great industry—the by-product industry connected with the destructive distillation of coal and wood.

#### TECHNICAL ELEMENTS OF BY-PRODUCT COLLECTION.

When substances such as coal, wood, and petroleum are destructively distilled, each particle or molecule is broken up into two or more molecules. As the temperature is raised these may in turn be broken into still smaller molecules, until the original substance is resolved into its



ultimate elements, which are predominantly hydrogen and carbon, with relatively small proportions of oxygen and still less of nitrogen and other elements. At any stage of distillation the whole range of possible products may be in process of formation, although the products being formed at any time in greatest proportion depend upon the average temperature at that time.

The effect of primary distillation may be such that some or all of the products are carried to places where the temperature is higher than it was at the point of decomposition. In this case certain of the products may be broken up two or even more times before finally leaving the retort.

For example, products of distillation from the interior of a piece of coal have to pass through outer layers of coal where the temperature is higher than at the point of decomposition. Or, again, the volatile products may pass through a region of higher temperature in the retort above the charge. The products of any stage of distillation, therefore, will consist of numerous chemical combinations resulting from various primary and secondary distillations that have taken place, at widely differing temperatures, at different points in the retort simultaneously.

It is almost invariably found that one or more of the products of destructive distillation are of such a composition as to remain in a gaseous condition at ordinary temperatures. These are usually spoken of as gases, or permanent gases; although some of them may be condensable to liquid at temperatures only slightly lower than those to which they are subjected in practice.

The stream of gas coming ordinarily from the retort is partly composed of compounds that are later to be condensed to liquids. In all cases, also, a solid residuum, the nature of which depends upon the temperature attained in the retort, remains behind. If the temperature is high enough, the residuum will be of carbon and ash, the temperature required to obtain this result depending upon the nature of the substance being distilled. The problems to be dealt with are those involved in the collection and further treatment of the matter carried from the retort in the stream of gas.

In every part of the retort the mechanical action is such that the emerging stream of gas carries from it suspended particles of liquids in the form of fog, or of solids in the form of smoke. As the vapours are cooled certain gases condense to liquids, either from contact with a

cold surface, or in the body of the gas itself. In the latter case, either new fog particles will be formed, or they will be absorbed in solution in fog particles already present.

It should be particularly noted here that, for the purposes of this paper, a substance is spoken of as a "vapour" when it is incorporated invisibly in the gaseous products, but is condensable to a liquid condition at lower temperatures used in the industry. The term is not applied to fog particles. The word "gas" is so defined as to include those substances that remain in the gaseous condition at all temperatures used in the industry. "Condensation" is defined as connoting all transitions from vapour to liquid, whether the liquid forms directly on a cooling surface or in the body of the gas as fog.

The products of destructive distillation may be separated for the sake of convenience into two subdivisions—to wit, that which remains as gas at the close of the operation and the part that has been condensed and collected as liquid. The condensable constituents ordinarily divide themselves more or less perfectly by settling into two distinct portions or fractions, one of tar or oil, and the other of an aqueous appearance.

Destructive distillation takes place practically always at temperatures above the boiling point of water. Hence the smoke, or fog, which is carried in the vapour stream from the retort, consists of particles of carbon or of tar fog which do not volatilize at the temperature of the vapour stream. As cooling takes place, further portions of the same tarry substances condense, as may also some of the lighter tarry or oily constituents which are wholly in the form of vapour at higher temperatures. These lighter tarry and oily compounds do not form new fog particles to any appreciable extent; they are simply absorbed into the fog particles of heavier tar already carried in suspension in the vapour stream.

As the temperature of the gas drops to a certain point, water vapour begins to condense, the temperature of condensation depending upon the relative amount of water vapour present in the gas. The condensing water carries with it in solution certain of the distillation products that are soluble in water and not soluble in tar or oil. The most important products thus obtained in solution in the condensed water are ammonia from coal distillation, and acetic acid and wood alcohol from wood.

As far as the writer has been able to determine, it is very doubtful if any appreciable amount of water fog is ever formed in the externally cooled condensers used in the distillation of coal or wood or oil, although it may be formed quite freely if the cooling of the gas is effected by a spray of water injected into the gas. As the gas is lowered in temperature by contact with a cooler surface, condensation of water vapour is effected. It is a property of water that it will adhere immediately to that surface. If water fog is formed, it readily collects against even an oily or a tarry surface. Thus one may collect water from gas passing through a condenser with cooled walls, even though the gas itself emerges from the condenser at a temperature much higher than that at which water fog is formed.

On the other hand, tar fog possesses a unique physical property that has determined the whole course of the by-product industry. It is extremely difficult, if not impossible, to collect tar fog from a gas by driving it against baffling surfaces when the temperature is high. It is true that a part of the fog can be collected by driving it violently against a solid wall, using for this purpose a rapidly revolving centrifugal blower; but the removal is far from complete.

When, however, the gas is cooled to temperatures well under 50°C. (122°F.), the tar fog readily adheres to the surface of contact; but even at this low temperature there are occasionally very small proportions of the fog that remain persistently in the gas. This residual tar may go through the purifying processes of a city gas plant, and even through a benzol recovery apparatus, and finally fall in the holders or distributing mains. So difficult of removal is it, that it may be driven through a closely woven filter paper without causing any appreciable discolouration.

The cooling of the gas is not only a very expensive proceeding, especially where it is not practicable to obtain water at a temperature much below that to which the gas must be brought, but it also involves a much more undesirable feature. In all coal and wood distillation, as at present carried on, water vapour condenses to some extent at least. Hence in collecting the tar it is also necessary to condense and collect a water fraction.

The ideal of those engaged in the distillation of coal and wood has always been to devise some means whereby the gas could be cleaned of tar fog at any and all temperatures above the condensation point of water vapour. This would make it possible to collect the tar fraction-

ally and free of water. Further, there might be a direct collection of ammonia in the form of ammonium sulphate, and of acetic acid as acetate of lime, without any previous condensation and re-distillation of water. The work described in the following pages was undertaken in the hope of demonstrating the possibility of achieving this ideal.

All the experiments conducted by the writer were conducted on what corresponds to a commercial scale. The process of electrical precipitation was tried on so many distillation gases, at such great extremes of temperature, and with such marked success, that it may be confidently asserted that an ideal method of collecting by-products is now available.

Electrical precipitation has been used with success in various places for the purpose of collecting residual tar fog after the gases have been through the ordinary condensing and scrubbing systems. So far as the writer is aware, however, the work here outlined constitutes the only successful attempts to demonstrate the efficiency of the process in collecting all of the tar at high temperatures.

#### EXPERIMENTAL WORK IN ELECTRICAL PRECIPITATION OF TAR.

Tar fog is known to be very easily precipitated. Also it always collects on the treater pipe in liquid form, and runs out without sticking to the pipe or clogging it in any way; although if collected at a high temperature it may harden or become viscid after cooling. The power consumption is exceedingly small in all cases that have been investigated by the writer or that have come to his attention.

The installations for collecting tar will always be very small in comparison with those needed, for example, in cleaning smelter or cement kiln gases. In the distillation of coal approximately 10,000 cubic feet of gas per ton is evolved. Hence, even a large distillation plant does not produce more than a very few million cubic feet of gas per day; whereas an ordinary smelter or cement-plant stack will carry off hundreds of thousands of cubic feet of gas per minute. A single treater pipe is expected to de-tar completely a million cubic feet of gas per day. Thus a very few pipes are sufficient for an ordinary installation; whereas in one smelter in the Northwest the installation consists of about fifteen hundred pipes. The engineering problems involved in tar collection are, therefore, inconsiderable, the one imperative condition being that the plant should be gas tight.

An outstanding difficulty, however, was encountered, at the very beginning of the work. Reference to figs. I, II, and III shows that the

insulators A and B may become covered with a layer of tar. This may destroy the insulation completely, or it may cause the leakage of so much electricity from the wire over the insulator to the pipe (instead of through the gas) that a short-circuit will be produced. Or the leakage may be so great as to cause a serious loss of power. Such a loss may so reduce the capacity of the high potential transformer that the requisite voltage can no longer be maintained.

Two general lines of approach have been used in attacking this problem. One possible solution may lie in devising such a treater as would prevent the formation of a layer of tar on the insulators; the other is the development of an insulator that will not permit of any serious loss of power or lowering of treater voltage even when covered with tar.

The firm of Sir Oliver Lodge and Sons, Birmingham, seem to have been the first to attack the problem in the former of the two above-mentioned ways. The history of their attempts is not made very clear in the published records; but they appear to have finally taken refuge in efforts to prevent condensation of tarry matter in the electrodes by means of heating coils within the electrode itself.

The experience of the writer suggests that this may in certain cases be of assistance in preventing the condensation of water upon the insulator; but it would not prevent the deposition of tar. Although the tar would then be distilled by the heat from the interior of the insulator, there would be left behind a layer of pitch or of pure carbon were the temperature carried high enough. This layer would ultimately become more conductive than a layer of ordinary tar.

Since the work was not continued in England, the writer assumed that the experiment had been unsuccessful, and that accordingly nothing further had been done.

Fig. II represents the type of treater developed by the writer. It embodies devices to protect the insulator from becoming contaminated with tarry matter. It will be noted that the insulators are removed from the main current of the gas; hence fog or smoke can be carried to their surfaces only by eddies or convection currents in the gas. It will also be noted that there is a nipple of treater pipe between each condenser and the main current of gas. This is designed to precipitate fog or smoke from the eddies of gas and thus protect the insulator. Such devices have been used in plants of various kinds, notably in the plant of the Minnesota Steel Company, Duluth, Minn. At this plant

the residual tar fog is removed from producer gas after it has gone through a standard scrubbing and washing system. This installation has been working for a considerable time with complete satisfaction. The writer would point out, however, that the precipitation of these small proportions of very finely divided residual tar from producer gas is exceedingly easy as compared with the collection of the total tar content from such gases before they have been cooled.

Just before the writer's work was begun, Professor White, of the University of Michigan, completed an excellent piece of investigation, although the results of his work were not published until a month or two later.<sup>1</sup> Professor White used a rigid discharge electrode, held in the middle of the pipe by a rigidly supported upper insulator, thus doing away with the lower insulator as shown in the figures given. His idea was that the gas would be completely cleaned before reaching the insulator, which could not then become contaminated by deposition of tar while the treater was working properly.

It may be remarked that not only would it be obviously necessary to clean the insulator after each momentary or partial failure of the treater action from any cause, but that also it is not at all probable that absolutely all the tar could be removed continuously for any length of time by any treater working even at maximum efficiency. Professor White succeeded in de-tarring approximately half a million cubic feet of gas per day in the plant of the Ann Arbor Gas Company, but did not solve the problem arising from periodical contamination of his insulators. Thus his experiment was hardly a commercial demonstration. In passing it may be stated that his paper includes a very clear résumé of the advantages resulting from de-tarring coal gas at various temperatures.

Incidentally, mention may be made of the fact that the treater pipe at Ann Arbor was placed in the line of gas after it had passed through P and A tar extractors; hence the gas was effectively cooled and cleaned before reaching the treater. The problem presented was, therefore, the easiest in the whole range of tar precipitation.

Mr. Steer, who assisted Professor White, then undertook to investigate the de-tarring of illuminating gas in his own way. It is known that the residual tar in gas after passing through condensing and scrubbing systems, consists of a very fine fog or fume, which is so

<sup>1</sup> The Electrical Separation of Tar from Coal Gas, Second paper. Alfred H. White, R. B. Rowley and C. K. Wirth, 23rd Annual Meeting, Michigan Gas Association. September 2-3-4, 1914.

light that it is difficult to force it effectively against any baffling surface. Mr. Steer made use of the well-known fact that a discharge of electricity into gas will ionize it and that ionization promotes the agglomeration of the fog particles into larger drops which are more easily removed from the gas. He employed an alternating current for his discharge, and hence did not use electrical precipitation in the accepted sense. Whilst Mr. Steer's installation treated residual tar satisfactorily, so far as the writer is aware he did not undertake to solve the problem of total tar removal at higher temperatures. As a matter of fact, the wider problem cannot be met in this manner; Mr. Steer's application is simply an effective auxiliary to present washing and scrubbing systems.

It is appropriate to note here that electrical precipitation (and therefore the Cottrell Process) connotes much more than the field covered by Mr. Steer.

#### THE WRITER'S EXPERIMENTAL WORK.

##### *Work at the Plant of the Vancouver Gas Company.*

In the mid-summer of 1914, experimental work was commenced at the plant of the Vancouver Gas Company, under an arrangement made with Major R. W. Ford, the company's manager. The company undertook to supply the necessary labour and the material for the treater pipe. The first treater was installed in a 6-inch line leading from a gas producer burning bituminous coal. The gas was heated, without washing or scrubbing, under one bank of horizontal retorts. Hence the experiments at this point did not interfere with even the work of this subsidiary unit. The gas passed through the treater pipe at temperatures depending upon the stage of the charge in the producer, but usually remained under the boiling point of water. The action of the producer was varied at different times in order to vary the gas temperatures, the amounts of suspended tar, or the amounts of undecomposed steam.

The treater pipe, used here and in the subsequent work at this plant, is shown diagrammatically in fig. I. It will be seen that the gas enters at the top of the treater pipe in such a manner that the upper insulator is removed from the direct impact of the tarry gas. No particular attempt was made to protect the lower insulator, because it was expected that the action of the treater in cleaning the gas would prevent its contamination. The function of the lower insulator (figs.

I and III) consists simply in preventing the wire used as a discharge electrode from swinging out of the centre of the pipe. In these two designs the wire is kept taut by a weight suspended from the bottom. In fig. II the lower insulator serves to maintain the tension in the wire as well as to prevent swinging.

Although, theoretically, all the suspended material in the gas is driven to the collecting electrode, which in this case is the pipe, yet always more or less collects upon the discharge electrode. In fig. II this tarry material runs down over the lower insulator, which, accordingly, must maintain its insulating properties when covered with tar, if the treater is to operate. It will be observed that the lower insulator is protected from this action in the types shown in figs. I and III. A weakness of the arrangement shown in fig. I, as compared with that in fig. III, appeared in the course of the work; although, for due cause, no change was made at the time.

As the temperature of the gas fell from time to time, there was always a considerable condensation of vapours of water and light oil to the form of fog. Some of this, as well as some of the heavy fog that had escaped collection, would necessarily collect upon the lower insulator, and would gather as drops along the length of its lower side. Thus a body of liquid would be formed, more substantial than the thin layer found over the upper portions of the surface, and hence greater electrical leakage would result. This leakage did on occasions carbonize some of the condensed matter on the lower side of the insulator causing a breakdown in the insulation that could be remedied as described later. This difficulty did not occur frequently and the serious task of changing the lower part of the treater pipe to conform with fig. III was not undertaken.

Attention was concentrated upon providing satisfactory devices for an upper insulator as shown in fig. I. No treater is absolutely perfect. There will always be occasions when, for one reason or another, performance will not be normal for longer or shorter periods. Hence, all treaters must be designed to take care of a possible deposit of tar on the insulator.

Two devices were tried with the hope of lengthening the period of effective action of an insulator indefinitely. Cleaned gas from the holders was driven into the upper insulator chamber through the opening shown at M, to find out if the dirty gas would reach the surface of the insulator, even if it should by any means get through the



nipple into the insulating chamber. Later, also, silk bags were tied between the walls of the pipe and the insulator (as shown at E), with the object of increasing the effectiveness of the clean gas introduced through M, and also with the idea that the distribution of the clean gas would be such that eddies of clean gas from below would be more completely prevented. In this case the silk bag became essentially the insulator, or, at least, an integral part of it.

These two devices were effective in retarding the formation of the layer of tar over the upper insulator; but we were finally convinced that they would not be satisfactory. A window was then placed in the top of a treater pipe through which the action of the gas could be studied with the aid of a beam of light thrown down into the insulator chamber. In the absence of the silk bag, little streams of tar-laden gas could be seen occasionally to reach up into the chamber against the flow of clean gas. Some of these were seen to deposit tar upon the insulator. When the silk bag was used, it was darkened in spots by the deposition of small quantities of tar, until, finally, a path for a small leakage of current was produced. The bag would then be destroyed almost instantly. These reactions occurred against a stream of clean gas equivalent to the total flow being handled by the treater. Were it possible to make the flow of clean and dirty gas entirely uniform, and to secure unvarying electrical discharge, the devices in question would meet the case; but these conditions are not attainable.

After much work in the directions indicated above, the conclusion was reached that it might be possible to find an insulator that would operate even when covered with a layer of tar. Tarry insulators had already worked for considerable periods. Experiments showed that an insulating length of a couple of feet would not entail a serious loss of power at the requisite voltages when the insulating surface was covered with as thick a layer of clean tar as could be made to adhere to it. The ability to insulate was not destroyed even when the tar contained as much water as it could readily hold in suspension.

On the other hand, one foot of insulating length would allow a leakage of current that would quickly develop into a flaming discharge, carbonizing the tar and leaving a permanent conductor behind. Hence it was decided that an insulating bushing, reaching three feet inside the insulating chamber, and far enough outside to prevent sparks from the wire to the pipe (as between R and S in fig. I), might stand the duty without any protection whatsoever. Bushings made of many different

kinds of insulating material were then tried. All of these lasted for a considerable time; but all, with one exception, were destroyed at the first breakdown of the insulation.

It appears that at C, in fig. I, there is a continuous brush discharge reaching out from the wire and upward over the tarry surface for a few inches. This discharge seems to evolve heat enough to carbonize the tar very slowly. If the tarry layer is not renewed or rapidly removed, a residue of carbon will form upon the surface. Being a conductor, this will permit the brush discharge to creep up the insulator until a line of carbon is formed from C to S. A flaming discharge forms along this line at a voltage depending, apparently, upon the kind of tar being handled in the treater. The voltage at which this phenomenon occurs is not high enough for efficient precipitation in the treater pipes. An attempt to raise the voltage would simply result in greater current consumption.

Professor White seems to have been troubled in the same way, for he reports that his insulators would break down after a few hours, and recover when the layer of tar was burned off. The writer, however, has never been able to burn off this carbonized line, although in many cases heavy currents have been driven over the insulator for hours at a time. However, in a report of later work on a larger scale, Professor White states that it was necessary to shut down a treater every few hours in order to clean the insulator. He does not state that it was impossible to burn off the layer of tar with the insulator in place; but presumably that was his experience.

The flaming discharge over a carbonized line will, in a very short time, cut into composite insulators such as bakelite, micanite, etc. Insulators of earthenware, porcelain, etc., either cracked throughout their length, or the glazing broke under the line of discharge. Carbon would then be deposited in the porous material beneath, and would remain as a permanent conductor. If the power capacity of the transformer used were very small, as was the case with Professor White, the destruction of the insulators might not be complete when the carbonized line was formed; but it happened in every case with the larger transformers used by the writer. Insulators of various types were tried, and, although all were destroyed in turn, yet the results were so promising in every case that the search for the proper insulating material was continued. There was no difficulty at any time, while the insulator was still in condition, in cleaning all the gas that could be removed to the

treater by the producer. Pitot tube readings indicated that at times gas was delivered at the rate of almost half a million cubic feet per day.

Owing to changes in the plant it was necessary to alter the location of the treater while the search for the proper insulator was still in progress. It was then installed to handle the gas from the carburetted water gas plant in which raw coal was used instead of coke. The gas was taken from the top of the first water seal just outside the generator, with the intention that hot gas directly from the stand pipe would be taken if experiments with the lower temperatures were successful. The gas came through the treater at about the same temperature as did the producer gas previously used, and conditions were in all respects much the same.

Shortly after the treater was installed in its new position, a tube of fused silica was tried as an insulator. It did not take long to prove this the ideal sought for. Preliminary experiments, outside the treater pipe, showed that a flaming discharge over a coating of carbonized tar on the silica does not injure the silica in the least. It was impracticable to burn off the carbonized tar with the current within a reasonable time; but when it was removed by an intense flame, no trace was observed of a carbonized line from the previous treatment.

The first tubes available were only 30 inches in length; but preliminary experiments indicated that this length would suffice for fair service. The treater pipe was then rigged up with the new insulators in position. Gas was passed through the treater to drive out air and obviate the danger of explosion. The current was then turned on, and appearances indicated that the insulator was practically short-circuited. The treater was then opened up to see if anything had been broken, in assembling; but all was in proper order. It was assembled again and tested electrically with air in the treater pipe, and the insulators were found to be in perfect condition. Gas was again turned into the treater pipe, and the current started as before-with the same result. This was repeated a number of times, until finally it seemed that the fused silica would not remain an insulator, although it would recover its properties on exposure to the air.

There seemed to be no explanation of this remarkable phenomenon until, by accident, the current was left running for a considerable time. It was then noticed that, after about an hour, the insulation became gradually better, that is, the voltage kept going up and the current down, without any change in the setting of the controlling rheostats.

In two hours the insulation had completely recovered, and precipitation was proceeded with in a perfectly normal manner.

Initial failure and gradual recovery have been observed in every case where fused silica has been used as an insulator. A clue to this behaviour seems to have been found in the recent work at Sault Ste. Marie, as will be explained later. The recovery of the insulator, after its initial failure, takes different lengths of time, depending apparently upon the kind of tar being collected. In work on petroleum distillation described below, recovery was complete in a minute; while in hot carburetted water gas from the stand-pipe at Tacoma, the silica improved for the first hour, when it reached good condition, and then more slowly for as much as half a day.

The first experiments with short fused silica tubes at Vancouver showed that after the initial failure and recovery, the insulator remains perfect for periods varying from six hours up to twenty-four. The cause of failure is mentioned above. It appeared that longer insulators give entire satisfaction; but before these could be obtained, changes in the management of the plant made it impossible for the work to be continued. It had been carried on continuously during three months of the summer, and then at available intervals in university lecturing during the winter and spring of 1914-15.

#### *Work at the Plant of the Tacoma Gas Company.*

In January, 1916, arrangements for the continuation of the work were made with Mr. W. A. Leuenberger, Superintendent of the Tacoma Gas Company. The treater pipe shown in fig. II was built, and used with two different kinds of gas at various times during the first six months of the year. No attempt whatever was made to prevent the formation of a layer of tar on the insulator. Tar collected on the discharge wire would run down over the lower insulator; while tar not precipitated in the pipe would collect on the upper insulator.

c. The lower insulator was made of two small quartz rods, 18 inches in length, tied together as indicated; the upper was a double quartz tube, 4 feet long, with approximately 3 feet of its length inside the insulator chamber. The use of a tube is necessary to permit of the entrance of the electric current, and a double tube was used to prevent puncturing of the insulator between the wire and the top of the treater pipe at the point S. The concentric cylinder F, in the lower insulator chamber, was used to prevent the swinging of the lower insulator caused by

the irregular impact of the gases entering the treater; and also to prevent the deposition of particles of pitch directly from the gas on the maul bar. This latter deposit is more conductive than the more fluid tar running down the discharge wire.

The treater was first installed to handle coal gas before the gas had entered the P and A extractor, and after it had been through the exhaustor, which was, in this case, the first piece of apparatus through which the gas passed after it left the collecting main.

At first the treater was used simply as a by-pass, with 4-inch inlet and outlet pipes. Valves on the main line, and on the treater inlet and outlet, made it possible to vary the amount of gas passing through the treater up to a maximum of over 100,000 cubic feet per day. Later, 6-inch pipes to and from the treater were installed, so that the total production of gas, about 400,000 cubic feet per day, could be passed through the treater at a temperature of from 120°F. to 130°F. when the plant was running normally. On one occasion, however, the water in the collecting main was lowered as much as possible and the temperature forced up to 160°F. All the phenomena were the same at both temperatures.

Great difficulty was experienced in obtaining quartz tubes that were not broken in shipment, and that would not puncture at or near the point S; but after such tubes were obtained, the work of the treater was all that could be desired, at least up to the end of each trial when a carbonized line was formed. The failure of the insulation never took place more than once a day. It had been hoped that this would not occur with the longer insulators; but the difficulty was met in another way.

Experiment showed that it was possible to wash off the tarry deposit on the insulator by allowing light coal tar distillate to enter at M and run down over both insulators. This carried off deposits that were becoming thickened, but that were not yet actually carbonized. It was shown that by this simple act of attendance, once or twice a day at most, the action of the treater could be made continuous. If the washing were delayed until a carbonized line formed, the insulators would have to be removed and cleaned in a hot fire. The feasibility of continuous treater-pipe action was now taken as proved for the temperatures indicated.

As it was not practicable to obtain coal gas at higher temperatures, the use of one of the carburetted water-gas machines was secured. The

gas was taken from the stand-pipes to the treater through a 4-inch line, about 50 feet long. The temperature in the treater varied from 450°F. to slightly over 500°F. at different times and under different operating conditions. The operation of the treater was much the same as it had been in all the work to date, except that the initial failure and recovery of the insulation took a longer time than in the case of coal gas or producer gas. At high temperatures the tar fog particles could not contain as much light oils as they would at lower temperatures. The tar collected ran off the pipe freely, but would harden as a mixture of coal tar pitch and petroleum asphaltum on cooling.

Tests were made on both the coal gas and the carburetted water gas to see if the calorific value of the gas were raised by the removal of the tar. No improvement was found. This indicates that at temperatures as high as 160°F. the particles of coal tar fog absorb from the gas practically as much light oil as would be absorbed at usual temperatures of removal, namely, from 100°F. to 120°F. Improvement in the calorific value of carburetted water gas when the tar was removed at 500°F. was expected, but the following facts account for failure to obtain it: The coke used was quite soft and contained a considerable amount of residual pitch, which would yield only small amounts of light oils on being distilled. The petroleum used in the carburetter and superheater had been "topped" at a fairly high temperature, and therefore could not be expected to yield much light oil in any case. On cooling, small quantities of the cleaned gas gave light yields of oils of medium density, but practically nothing of the gravity of benzol or gasolene.

The arrangement of the plant was such that the coal gas tar was collected with that from the water-gas machines. Moreover, the ammonia and tar-collecting systems were contaminated with tar from ordinary operation. There was therefore no opportunity to determine the effect of total removal of tar from the gas upon the appearance of the ammonia liquor. It would probably have taken weeks of running with tar-free gas to clean out the pipes and scrubbers. However, the cooling of small quantities of the gas leaving the treater pipe showed that a clean ammonia liquor would be produced by this process. Only rough measurements were made on the efficiency of the treater as a tar collector; but these indicated that the gas leaving the treater was much more thoroughly de-tarred than the gas at the entrance to the purifiers after passing through all the condensing and scrubbing machinery.

## APPLICATIONS IN WOOD DISTILLATION.

### *General Considerations.*

It is advisable to give forthwith a very brief sketch of the processes used in the distillation of wood, in order that the value of this investigation may be more fully understood.

Resin and turpentine, as such, exist in soft woods, and are driven off by ordinary distillation or are obtained by solution in absorbing oils. No by-products are obtained from hardwood by ordinary, as distinct from destructive, distillation. At a temperature of about 200°C. destructive distillation of cellulose begins to take place, that is to say, the wood is decomposed by the action of the heat. The products are pitch, creosote, light oils, and pyroligneous acid, the last yielding water, acetic acid, and wood alcohol. Hardwood tar is of little value, although creosote is constantly growing more valuable. The greater part of the tar from hardwood distillation is burned as fuel under the retorts.

Softwood tars are very much more valuable. The recent demand for light pine oils for use in the flotation of ores has enhanced the value of the tars appreciably. Pyroligneous acid from soft woods contains, on an average, less than one-half the acetic acid yielded by hard woods, and one-quarter or less, as much wood alcohol. The values of these two products are fixed by the hardwood distillation industry, with the result that pyroligneous acid from softwood is usually thrown away.

In wood distillation the products are all collected together in a single system of condensers. The tar fraction is then separated as completely as practicable from the pyroligneous acid by settling. However, the separation is not at all complete. The tar fraction contains some pyroligneous acid, very finely divided as a kind of emulsion. In refining tar by secondary distillations, this emulsion must be broken up, a very difficult operation in the case of most softwood tars, because of frothing in the refining stills when the temperature reaches the boiling point of water.

The pyroligneous acid from both soft and hard woods is a very dark-brown or black liquor, the colour being due to the presence of so-called soluble tar. When the acid is evaporated this remains in the still as ordinary tar, and it cannot then be placed in suspension in the acid. Hence it is probable that the tar is present in the acid in the molecular state. The amounts of this soluble tar in the acid from

different kinds of wood vary greatly. In general it is much greater in acids from soft woods than in those from hard woods. Statements have been made to the writer to the effect that there may be as much as 12 gallons of soluble tar in the pyroligneous acid in a cord of some pine woods. Where the tar is valuable, as in the case of pine woods, the waste of the pyroligneous acid involves a serious loss of valuable tar.

#### *Experimental Work at Seattle.*

In the summer of 1916, the writer obtained the use of an experimental plant, belonging to the United States Forest Service and the University of Washington, situated on the grounds of the latter institution at Seattle. Work was carried on here with the co-operation of Professor Benson of the Department of Industrial Chemistry.

The plant is a complete model of a softwood distillation plant on a semi-commercial scale. It is composed of the retort, air condenser, water-cooled condenser, exhauster, and gas holder, with various accessories, such as stills for refining the tar and pyroligneous acid. It takes something over three-quarters of a cord of wood at one charge. Examination of the plant records shows that gases from the retort enter the air condenser at temperatures slightly above the boiling point of water. This indicated, as was proved later by experiments, that at this point the applicability of electrical precipitation to wood distillation could be best investigated.

The treater pipe (fig. II) that had been used in the work at the Tacoma gas plant, together with the experimental outfit used there, was brought to Seattle. The pipe was installed between the retort and the air condenser in such a way that all the vapour-laden gas was forced through the pipe without any by-pass. During a part of the run, after destructive distillation of the charge had begun to take place, the temperature of the treater remained slightly above the boiling point of water until the later stages of the run, when the quantity of gas was so small that radiation lowered the temperature below this point.

At such temperatures, water, acetic acid, and light oils were above their points of condensation, and thus went through the treater pipe as true vapours, without being in any way affected. Hence the smoke or fog consisted entirely of particles of heavy tar, which were completely removed during the passage of the gas through the treater. In these circumstances the material collected in the treater was a heavy anhydrous tar, which could be placed on the market directly as



"refined." The light oils and pyroligneous acid were then condensed and collected in the condensing system of the plant. In all the runs made at this plant, roughly selected fir mill waste was used. Each piece was supposed to show some sign of pitch, so that the large quantity of tar distilled would demonstrate whether its presence in the gas would make electrical insulation more difficult.

The total yield of tar averaged about 40 gallons per cord. Of this amount, from 10 to 12 gallons were collected in the condensers as a light oil. There was marked variation in the total and relative yields of tar and acid.

The light oil rose to the top of the pyroligneous acid almost immediately after condensation, and did not carry with it any appreciable proportion of pyroligneous acid as an emulsion. It now appeared that the soluble tar obtained in the regular processes of distillation consisted in great part of heavy tar and not of light oil. The heavy tar in our work was already removed in the treater, and, since the light oil could not remain in solution or suspension in the pyroligneous acid, the latter was found to be a very light straw-coloured liquid containing substantially less than 10 per cent of the amount of soluble tar found in the darker acid ordinarily distilled.

#### *Tests in Flotation of Ores.*

The light oils collected in these runs were tested with various ores of the Northwest to determine its suitability for use in concentration by flotation. It was found to possess substantially the same properties as the commercial grades of Southern Pine flotation oils. It may be pointed out here that a flotation oil should possess two distinctive properties: first, it should make an effective froth; and, second, there should be enough body to the froth to lift the valuable mineral constituents of the ore with the least practicable quantity of gangue. Light oils usually possess the greater frothing capacity; while heavy tar in the oil lends body to the froth.

Pine-wood distillation, carried on as in the above tests, yields a heavy tar and a light oil from the first operation, thus doing away with the ordinary secondary or refining distillations. The requisite proportion of light oil and heavy tar for a flotation oil suitable for a particular ore can be determined by experiment.

During the winter of 1916-17, Professor Benson and his associates drew up plans for a distillation plant for the production of flotation oils

The scheme was dropped temporarily on the entrance of the United States into the war. Nothing further has been done in applying electrical precipitation.

#### *Suggestions.*

There is no manner of doubt that a heavy wood tar can be collected by electrical precipitation at a temperature above the boiling point of water, without any of the difficulties in the way of insulation that have been encountered in collecting coal tars from horizontal retorts and from by-product coke ovens.

The fact that light oil vapours, which pass through the treater pipe with water vapour at high temperatures, do not, on condensation, remain in suspension in water to any appreciable extent, suggests that there may be now developed a direct process for the recovery of acetate of lime. If such a process were developed, it would constitute a revolution in the methods of distillation of both hard and soft woods.

Following up the suggestion of the previous paragraph, it seems commercially possible to collect by-products from charcoal kilns or gas producers burning wood. This is not done at the present time because of the cost of by-product collection equipment, and because considerable quantities of the valuable by-products are burned in the kilns and the remaining amounts would not warrant the investment necessary for their recovery. On the other hand, the expense of an electrical precipitation installation is merely nominal. This alone would yield considerable quantities of heavy tar. A direct process might then be made to yield sufficient acetate of lime to be worth recovery, without condensation. The recovery of either or both light oil and wood alcohol could be effected in the manner that might appear most desirable.

In the opinion of the writer, this is the only promising method of attacking the problem of utilizing economically the enormous quantities of wood waste in Western Canada and the Northwestern States.

#### APPLICATIONS IN PETROLEUM DISTILLATION.

Previous work in coal and wood distillation suggested new possibilities in the fractional distillation of tar, petroleum, and other liquid hydrocarbons. Mr. J. A. Cunningham, manager of the British Columbia Refining Company, Port Moody, B.C., authorized a series of tests at this plant in October, 1916. Visual examination of the vapours leaving the stills revealed the fact that there is carried over a consider-

able quantity of smoke or fog. It was with the idea of collecting this that the work was undertaken.

Gasolene is not produced at this plant. The crude petroleum is of a heavy gravity, and is all obtained from the Union Oil Company of California. The oil is evaporated until asphalt of the required degree of hardness is left in the stills. Small quantities of gasolene are yielded, but it is not separated from the lightest distillate.

The treater pipe that had been used at Tacoma and Seattle was sent to Port Moody and installed on the 6-inch line leading from one of the stills to its condenser. All, or a portion of, the vapour from the still could be passed through the treater. In the tests made all the vapour was handled. The regular charge in the still was 165 barrels. The temperature of the treater pipe rose from time to time during our runs until, at the end, with the asphalt remaining in the still, it stood in the vicinity of 450°F.

The advantages expected were based on three theoretical assumptions.

It is known that the distillation products from liquid hydrocarbons do not exist as chemical entities in the undistilled hydrocarbon. Distillation is destructive. It is doubtful if distillation is ever achieved without a certain amount of "cracking." The phenomenon of cracking is not at all clearly understood. However, it is reasonable to suppose that the breaking up of a molecule may give two or more molecules, each with a smaller number of atoms. The composition of the smaller molecules depends on the temperature at the moment of decomposition. In general, one at least of the resulting molecules will contain more hydrogen than the original molecule, and one at least will contain proportionately less. Ultimately, if the temperature is carried high enough, it is possible to crack the compound into hydrogen and carbon.

It is fair to suppose that the violence of the reaction when the molecule is broken up, will carry the products of decomposition into the body of the resulting vapours. This implies that the lighter molecules would be present as true vapours, while the heavier might agglomerate into fog particles, or even remain as suspended particles of carbon. All the heavy constituents would be carried in suspension with the stream of true vapour and collected in the condenser with the lighter fraction.

The second consideration involves the action of heat in causing true, as distinct from destructive, distillation. The stills used in refining tar, petroleum, etc., are heated by direct firing from below. At any given moment the body of the liquid in the still might have an average uniform temperature, though the temperature of the liquid in contact with the bottom of the still would naturally be higher. Since petroleum is viscous, particularly in the later stages of a run, when heavy engine oils are being driven off, it is probable that convection currents cannot be rapid enough to keep the liquid in contact with the hot bottom at a temperature as low as the average of the liquid above. There will thus be greater evaporation of the bottom liquid than could be produced at the average temperature of the still. This means that some of the heavy vapours rising from the hot bottom will be condensed to fog while passing through the cooler liquid above.

In the third place, the bubbling of vapour through the liquid is assumed to cause the formation of small particles of the liquid as a kind of spray. Such particles, if present at all, would not have been subject to evaporation, and would be carried over mechanically in the vapour stream to the condenser. A great deal of effort has been expended in attempts to extract these heavy fog particles and to drop them back in the still for proper fractionation. The ordinary dephlegmating column, however, achieves the desired result very imperfectly, because of the fact, mentioned before, that tar fog particles will not adhere to baffling surfaces satisfactorily at high temperatures. On the other hand, the electrical precipitation process proved a perfect dephlegmator; it was demonstrated that the vapour stream could, by this process, be made visibly clean of fog at any temperature reached during distillation.

It was expected that the tests would throw some light on the problem of "acid sludge." As is well known, the agitation of a fractional portion of tar or petroleum with sulphuric acid yields varying quantities of a dark heavy substance, which is of the consistency of a soft asphalt and is called "acid sludge." There is a difference of opinion as to whether this sludge is composed entirely of a chemical compound formed by the action of the acid on certain components of the oil; or whether a substantial proportion consists of asphaltic matter, carried over from the still mechanically and separated from the distillate by the action of the acid.

In the first tests the old treater pipe with soldered joints were welded with oxy-acetylene and used as an indicator of possibilities. No steps were taken to prevent surface condensation by insulation. The material collected in the treater was run into separate containing tanks and not back into the still. It was found that, as the distillation progressed, the liquid obtained from the treater was always about 7° Beaume higher than that collected in the condenser. The condenser liquor was run into the storage tanks with the corresponding material from the other stills that had not been through the treater pipe. Hence it was not possible in these first tests to make determinations on a large scale; but laboratory tests indicated that liquor obtained from vapours that had passed through the treater pipe would need considerably less acid and yield correspondingly smaller amounts of acid sludge than liquor of the same gravity produced ordinarily in the plant.

By far the most striking phenomenon observed was the presence in the liquid collected in the treater of very considerable quantities of free carbon, which settled in a soft granular condition. It is probable that this carbon was present in the vapour stream in the form of very small particles of carbon smoke, quite distinct from the tar fog particles. Probably, also, the particles were driven against the pipe by the electrical discharge in such a way that a layer of soft carbon was formed, and then washed off in granules by the streams of oil running down the pipe.

It was now determined to proceed with the erection of a well-designed treater pipe, thoroughly insulated to prevent surface condensation. It was also decided to install a more efficient electrical plant. These tasks were finished about the middle of February, 1917, and the investigation was resumed.

The results were wholly disappointing. Insulation did not increase the amount of treater liquor collected. No improvement over the former tests was noted. The absence of the granular carbon obtained in the earlier runs was attributable to the character of the oil, not to the process. However, the demonstration of the occurrence of free carbon in the vapour stream contributed something to our knowledge of oil refining. The possibility of sharpening the fractional cuts by the use of electric treater pipes as dephlegmators deserves immediate investigation.

#### WORK AT LOW TEMPERATURE DISTILLATION PLANT, NANAIMO, B.C.

In the early spring of 1917, an experimental plant was erected at Nanaimo in order to test, on a semi-commercial scale, the Thomas process for low temperature distillation of coal. The financial support was provided by the Merritt Collieries, Ltd., through the good offices of Mr. T. J. Smith, of Vancouver.

The method involves heating the retort by means of a return circulation of the gas produced in the process. The gas is heated outside the retort. In the experiment it was passed through heating coils placed in an ordinary fire box or combustion chamber. The heated gas was then led into the retort, and from the retort through a by-product collecting system before returning to the heating coils.

The by-product collecting apparatus consisted of an electrical precipitation treater pipe, two externally-cooled condensers, a scrubber filled with excelsior, a purifying box filled with chips saturated with sulphate of iron, and the exhaustor for maintaining the circulation. The piping was so arranged that any portion or all of the gas could be passed through the electrical treater pipe as it came from the retort, or from the first condenser in which it was partly cooled. The retort took a charge of about three tons. It was found that a maximum temperature of between 800°F. and 850°F. could be maintained, although in spots it reached as high as 950°F. The total volume of gas coming from the retort while a run was on, amounted to about 240,000 cubic feet per day.

The experiment failed to prove that the Thomas process is commercially profitable, although it did not prove that it is not.

Various causes led to the discontinuance of the work after a couple of months' trial. The amount of money needed had been very much underestimated, although double the amount first provided was expended. The company's excellent coking coal carried such a high percentage of volatile matter that, even at the low temperature of distillation, it would swell in the retort and prevent the circulation of the heating gas. The by-product collecting apparatus was built out of old material, and could not be made gas-tight, with the result that a large part of the by-products was lost. The amount of light oil found in the collected tar was disappointing.

The time needed to distil a charge of coal was greater than had been expected, and the heat needed to cause destructive distillation had been very much underestimated.

In spite of the failure of this particular experiment, the writer is of the opinion that the Thomas process may still prove valuable in special cases.

The main treater pipe, cleaning nipples, and upper insulating chamber of the treater pipe used are shown in fig. III, except that the nipples were only 6 inches long. The lower insulating chamber was as shown in fig. II, the gas going in at the bottom and out at the top. Hence, while the upper insulator was protected from the deposition of tar, the lower was continually covered with tar running down from the discharge wire. It was intended that a lower insulator chamber, shown in fig. III, would be used if that shown in fig. II proved to be a failure.

By arrangements of the valves, gas could be passed through the treater pipe at temperatures from  $380^{\circ}\text{F.}$  to  $160^{\circ}\text{F.}$ , and through this range it was found that a very non-conducting tar was produced. No sign of failure of the lower insulator was observed, although, in cases where a small amount of gas was passed through the treater pipe, the lower chamber would be so cold that a waxy deposit of tar oil with paraffin base, as much as a quarter of an inch thick, would adhere to it. Even at the highest temperatures at which the gas was passed through the treater pipe, as high as 90 per cent of the tar collected in the system was collected here.

It may, then, be confidently stated that electrical precipitation can be used for the collection of tar produced by the low-temperature distillation of coal without opening up the treater to clean the insulators, even though the treater is so constructed that the insulators are not protected from the formation of tar upon their surfaces.

#### \* WORK ON BEEHIVE COKE OVEN GASES.

In the spring of 1917, arrangements were made with Mr. Hewitt, of the Wilkeson Coal and Coke Company, Wilkeson, Wash., for tests on gases from one of their beehive ovens. The volatile content of their coal is considerably higher than is required for cooking. It was thought probable that a certain amount of by-products might be collected by specially working the ovens. It was proposed that, after the oven had been charged and air admitted for a sufficient length of time to develop coking temperatures, the air should be excluded for as long a time as practicable and the charge left to distil with the heat of the oven.

The trial failed because of a fact that had not been anticipated. Analyses of the gas coming from the oven, while the air was excluded, showed the presence of such large proportions of nitrogen that it was evident that enough air penetrated the walls and bottom of the oven to generate producer gas. It can be seen that partial combustion, and the very high temperature in the upper part of the oven above the charge, would destroy most of the tar and ammonia. No measurements of the amount of ammonia in the gas were made, because it was evident from the character of the tar recovered that most of the ammonia would be thus destroyed.

An open treater pipe, with the insulators outside, was used provisionally. It was to be replaced with a closed pipe of types I, II or III, should results warrant it. With this pipe the gas could be cleaned, as shown by ocular demonstration; but the problems of insulation, which are the chief difficulty, could not be studied. Tarry material, ranging from very thick tar to carbon with a small content of pitch, was collected at different times during the run. The most fluid tar obtained had a carbon content of about 40 per cent; while that produced during the latest stages of the run, when the oven and the upper portion of the charge were at their hottest, contained as much as 90 per cent. of free carbon. This did not run off the treater pipe or central electrode, but gathered as a kind of spongy mass that completely filled the pipe in a comparatively short time. It had the appearance of ordinary soot, but could be melted into a pitch which cracked open rather than flowed, even when warm.

It would be almost impossible to use electrical precipitation to collect this carbon. A small water-spray condenser was rigged up, and it was found that, with the lowering of the temperature, most of the free carbon smoke present in the gas could be washed out. In these circumstances, the smoke remaining in the gas was predominantly tar fog which probably could be satisfactorily collected by electrical precipitation, giving a clean producer gas for further use.

It was found that the opening in the door of the oven could be closed at intervals for a considerable portion of the run without lengthening the coking period, or interfering with the quality of the coke; but it is impossible to say whether this would apply to an oven that was air tight.

The trial gave only negative results, though it was promising enough to justify further investigation.



## WORK ON BY-PRODUCT COKE OVEN GASES.

### *Auspices.*

In the autumn of 1917, the Research Council undertook to support the continuance of this investigation in the one major field where no experiments had been made, namely, the collection of tar from by-product coke gases. It was advisable that work supported by the Research Council be done in Canada. This narrowed the location of the work to one of two Canadian plants, namely, those at Sault Ste. Marie, Ont., and at Sydney, N.S. Most satisfactory arrangements were made for the purpose at the former of these plants.

### *Note on By-product Oven Practice.*

The by-product coke oven consists of an oven charged with coal and heated externally to a high enough temperature to give a metallurgical coke without the admission of air to the ovens. Admission of air, through leakage or otherwise, involves the burning or cracking of tar and the destruction of the ammonia by-product. The gas from the coal, with its load of ammonia and of suspended tar fog, is led from each oven into a collecting main and from this into a by-product recovery house.

Modern ovens are built in banks, with heating chambers and flues below and between the ovens. Ovens may be of any width that gives a practicable coking time, though recent practice fixes the limit somewhere between 17 and 24 inches, giving a coking period of from eighteen to twenty-four hours. The temperature of the heating flues is regulated to give a coke strong enough for metallurgical purposes; 2,200°F. is ordinarily taken as the standard temperature. The collecting mains must be flushed with tar or with a mixture of tar and ammonia liquor, in order to prevent the deposition of pitch from the hot gases. The problem of keeping the mains clear is one of the chief difficulties to be contended with. By-product coke ovens yield from 6 to 7 Imperial gallons of tar, and from 20 to 22 pounds of ammonium sulphate per ton of coal.

### *By-product Recovery Plants.*

By-product recovery plants are of two general classes namely, those in which all the ammonia is recovered from the gas dissolved in water, and those in which it is carried by the gas into contact with a solution of sulphuric acid, with which it combines to form ammonium sulphate.

The latter are called direct-process recovery plants. Both of the Canadian plants are of the latter class, and it is in these that much may be expected of electrical precipitation.

As pointed out above, the gas coming from the ovens must be cooled to a temperature of approximately 104°F. to permit of the extraction of the tar by the methods in use. Cooling involves the condensation of a certain amount of water. In the first class of plants mentioned above, a sufficient extra amount of water is added to the system, in the ammonia scrubbers, etc., to absorb all the ammonia from the gas. In such plants the only benefits accruing from electrical precipitation is in giving cleaner ammonia water for subsequent purposes. The advantages to be derived from them will appear later.

#### *Direct Process Ammonia Recovery Plants.*

Reference to fig. IV will give a general idea of the construction and arrangement of a direct-process ammonia recovery plant. The figure is copied, with minor modifications, from a paper by T. V. Salt on "The Manufacture of By-Product Coke," presented before the Western Society of Engineers, October 6, 1916. The gas comes from the ovens at a temperature slightly below the boiling point of water, and the coolers must be of sufficient capacity to lower the temperature to about 40°C., though 30°C. is much more desirable if it can be attained. The condensed matter collected in the coolers consists of a small portion of tar with a considerable amount of very dirty ammonia liquor. As the gas reaches the cooler, practically all of the tar is condensed to fog particles. Cooling is necessary for the reason that it is impossible to collect the fog by mechanical means at such temperatures.

The temperature of the gas as it reaches the ovens is the dew point of the water vapour contained; but cooling to 40°C. brings it considerably below this point, involving its condensation in considerable quantities, which are collected against the walls of the system. Very little water fog, therefore, is carried with the gas. A certain amount of water will also be collected in the exhausters, and a considerable quantity with the tar in the tar extractors.

All of this condensable matter goes to the ammonia-liquor and tar-separating tanks. The ammonia is partly in the form of a solution of ammonia in water, and partly in the form of dissolved ammonium salts. Some of the ammonia can be driven from the liquor by heating it to the temperature of steam. The remainder, or "fixed" ammonia, must be

removed by displacement with lime. The ammonia vapour driven from the liquor in the still may be carried back into the stream of gas at any point, not necessarily to a point between the re-heater and the saturator as shown in the figure.

The function of the re-heater is a matter of controversy. Presumably it is intended to prevent the condensation of water vapour in the saturator; though it is not clear that this is necessary, because the heat of combination of ammonia and sulphuric acid is sufficient to keep the temperature of the saturator well above the temperature of the gas passing through it. Full descriptions of such a plant, particularly of the action of the saturator with its centrifugal drier, may be easily obtained by those interested.

#### ELECTRICAL PRECIPITATION AND DIRECT PROCESS PLANTS.

It was thought at the time the experiments were undertaken that a marked simplification of the expensive standard equipment might be made possible through the use of electrical precipitation. The results show that these expectations were justified. The chief equipment in the by-product house need consist only of an electrical treater installation, the exhaustor, and the saturator; thus replacing the coolers, tar extractors, and re-heaters by the treater pipes. It is considered practicable by the writer to insulate the treater pipes and prevent the collection of any water whatever with the tar, all the water vapour passing with the gas through the saturator without condensation. If the heat of the chemical reaction in the saturator is not sufficient to prevent the condensation of water vapour at this point, it is possible to raise the temperature a few degrees, by, for example, the use of a few steam coils.

In the experiments recorded later no attempt was made to insulate the main treater pipes, with the result that a small amount of ammonia liquor formed in contact with the pipe and ran out with the precipitated tar, though the average temperature of the gas remained distinctly above the dew point of the water vapour. This small portion of ammonia would go with the tar to a storage tank; but as there would not be sufficient of this liquor in the mains to flow, clear water would have to be introduced to assist in the flushing. Hence the ammonia liquor storage tanks could be made very much smaller, and the ammonia stills, with their auxiliary equipment, eliminated altogether. Such a recovery plant would be very much simpler in installation, operation, and renewal costs than those at present used.

#### *Agreement Covering Work at Sault Ste. Marie.*

It was decided in November, 1917, that a test of the methods adverted to above should be made at the by-product coke ovens of the Algoma Steel Corporation at Sault Ste. Marie, Ont. The Advisory Council agreed to meet the necessary outlay. Only a small amount of apparatus had to be purchased, the greater part being supplied by the writer, or loaned by the Western Precipitation Company, of Los Angeles, Cal. The other expenses, such as the purchase of materials actually used, wages, salaries, and transportation charges, were paid by the Council. Special appreciation must be expressed of the interest taken in the work by Mr. Fernsler, Superintendent of the Coke Ovens; by Mr. Duncan, his consulting engineer; and by Mr. Seymour, Mr. Fernsler's successor, who was in office for the last few days of the work.

#### *Position of Treater Pipe in Plant.*

The by-product house at Sault Ste. Marie contains, in parallel, three large coolers, four exhausters, three tar extractors, three reheaters, and three saturators. Inlet and outlet pipes from each unit go into main headers. There are no valves in the headers on the inlet side of the coolers, or between the coolers and the exhausters. The working of the plant was such that two coolers handled the gas, and the third could be used as a spare while one of the others was being steamed out to remove the tar. Similarly, one of the saturators could be used as a spare, while at times only one of the tar extractors would give as good service as two or three.

Without going into details, it may be stated that it was found impossible to devise a plan by which a given quantity of the gas could be passed through the treater pipe and into the saturator for treatment by itself. Accordingly, gas was taken from the main coming from the ovens, led through the treater pipe and one of the coolers (which was reserved for the purpose of our experiment, except during the steaming out period of about six hours once a week), and from the cooler into one of the exhausters between its main inlet valve and the exhauster itself. The exhauster used continuously a portion of the main current of gas. The amount drawn through the treater pipe could be adjusted by manipulation of the various valves. Gas that had been through the treater pipe was always cooled before passing into the exhauster, because it was found that the latter would pound if made to handle gas at too high a temperature. This made it impossible for us to observe

the effect on the appearance of ammonium sulphate produced from gas that had been de-tarred without lowering the temperature; but it gave the compensating advantage of enabling us to determine the effect on ammonia water collected in the cooler.

#### *Condition of Ovens.*

For some time the ovens at Saulx Ste. Marie have been in very bad condition. This has been due to various causes, but principally to poor labour and to the overwhelming demand for coke for iron and steel production. In keeping the ovens at their maximum coke capacity, the by-product output was necessarily sacrificed to a considerable extent. The oven became more and more leaky, and the mains were clogged with pitch. The pipes and valves from the ovens to the collecting chamber were also in very bad shape. The suction in some of the ovens was so reduced as to cause constant partial combustion of the by-products, which was continually aggravated by fires in the mains. The amount of air that found its way into the system is indicated by the fact that the gas contained much more than twice as much nitrogen as would be found in gas from a plant working in what would ordinarily be considered the worst shape.

These conditions were canvassed thoroughly before the experiment was begun, and, indeed, formed an added inducement as success here would assuredly mean success at other plants not so heavily handicapped. However, the obstacles in the path were by no means appreciated until near the close of the work. During the last few days of the experiment, a new main was installed on one of the two banks of ovens. The investigation was discontinued because of the construction of twenty-five new ovens.

#### *Power.*

After the treater had been installed, and the work started, it was discovered that the supply of power was inadequate. Current was obtained, not from the plant, but from the wires of the Great Lakes Power Company. The supply line also fed a number of motors in neighbouring plants, and could not be depended upon for regular service. The line voltage was led directly to the high potential transformer, and the rectifier on the high potential line was operated by a synchronous motor.

It was necessary to maintain the potential of the discharge electrode in the treater pipe very nearly as high as will give a spark from the wire to the pipe. As the voltage kept dropping from time to time, through as much as 25 per cent of its maximum, normal conditions could not be maintained. Remedy was sought in the employment of the motor-generator-rectifier set ordinarily used in the Cottrell process. This, it was found, followed the frequency and not the voltage of the line. At about the time this set was ready for use, one of the generating stations of the Power Company burned down. The remainder of the plant was overloaded and irregular in its operation during the remainder of the experiment. This induced so great a variation in frequency that we had to resort to hand regulation when the volume of gas through the treater pipe approached maximum capacity.

#### *Precipitation Phenomena.*

No attempt at a chronological record of work performed, nor detailed description of phenomena observed, will be made in this report. Attention will be confined to indicating the general results of the application of electrical precipitation to the de-tarring of oven gases.

The treater pipe, upper and lower nipples, and upper insulator chamber, as shown in fig. III, were used throughout the work. A lower insulator box, similar to that shown in fig. II, was given an initial trial. Previous experience showed that the difficulties of insulation increase, either with the temperature of distillation or with the amounts of free carbon or pitch in the tar collected. When temperatures of distillation are low, there is no evidence that insulators need special periodical cleaning; but cleaning was found necessary in gas from horizontal retorts, or from water gas plants where temperatures are lower than those in by-product coke oven plants.

The free carbon in coke-oven tar varies greatly, but in general is lower than that found in horizontal retort tar; although the proportion of pitch is higher in oven tar, where the temperature of distillation is 500° to 600°F. above that obtaining in horizontal-retort practice. In the lower insulator chamber, shown in fig. II, the insulator is continually covered with a layer of tar running down its surface from the discharge electrode. It was found that the failure and gradual recovery of insulation took place entirely as in previous work; but before the insulation had become normal, the carbonized lines would form as

described above. After thorough investigation, it was concluded that the tar in this plant is of such a nature that the insulators must be protected from deposition as completely as possible. Accordingly, a lower insulator chamber, as shown in fig. III, was installed and the work proceeded with.

Considerable work was done on the failure and gradual recovery of the quartz insulator. It was found that this phenomenon always occurred when warm tarry gas was admitted to a cold treater. The outstanding case was that in which the current was running with perfect insulation in cold clean gas when the warm tarry gas was admitted. The insulation became very poor immediately, and then recovered slowly during an hour or more. It was finally discovered that the insulation would remain perfect if the insulator were warm when the warm tarry gas was admitted. This discovery removes the only grave weakness that had been found in the earlier work. It seems probable that the condensation of tar and water on the *cold* insulator gives a partially conducting layer which may be destroyed gradually by the leakage of electricity which it causes.

This was the first time that the cleaning nipples and lower insulator chamber, as shown in fig. III, had been used. When operating conditions were such that the de-tarring was satisfactory, a new phenomenon developed. At varying intervals of an hour or more there would occur sudden increases of current, accompanied by noises in the rectifier. This was found to be due to the condensation of water on the insulators, giving a conducting surface which was destroyed quickly by the heat of the flashing electrical discharge across it. By far the greater amount of condensation takes place on the walls of the treater pipe, but there is a gradual deposition on the insulator as it cools when removed from the main stream of gas, as in this type of treater pipe. Presumably this would not take place if the temperature of the gas were a little farther removed from the dew point of water vapour. The "flashing" can be eliminated by an occasional heating of the insulator. To accomplish this, the insulator chambers are surrounded by a coil through which a small quantity of steam can be passed. The slight difference of temperature between the steam and the gas causes sufficient heating of the insulator.

Whether this result would obtain with other moist gases with this type of treater is not known; it was not observed in previous work on other gases with treaters of types I and II. Attempts were made to

prevent the "flashing" of the current by oiling the insulator, the idea being that the condensed water would collect in drops and not in a layer. It was found, however, that all the oils and greases tried would evaporate and leave the insulator unaffected.

Difficulty was experienced with the breakage of wires used as discharge electrodes. The breaks in every case occurred where the wire joined its support, either at top or bottom; and the only prevention discovered was to make the region of contact between the wire and its supports non-conducting. No explanation of the phenomenon is offered. Bright steel wire used as a discharge electrode would not tarnish. The tar gathering on it afforded protection from corrosion. Wire breakage did not occur previously except in dealing with petroleum vapours, and at that time no method of prevention had been discovered.

It had been hoped that by the use of this type of treater, insulators would give continuous service for an indefinite time without cleaning. This was achieved so far as the tar fog itself is concerned. An insulator could easily be made to run for a week or more without collecting a layer of tar on its surface; but a difficulty still remained. After a few hours running, a light, uniform layer of dry carbon was found on the insulators. In a short time this brought about a loss of current, and a gradual recovery to normal. After this rise and fall had continued for some time, the carbon layer became crossed in all directions by a net work of fine lines. The periods of increase and decrease varied with the amount of gas passing through the treater, and with the state of the ovens during the installation of the new collecting mains. It was never found possible to prevent this deposition of carbon on the insulators.

It is to be noted that carbon smoke is about the most difficult substance to collect with complete efficiency by electrical precipitation; whilst tar fog is amongst the easiest. The writer doubts if any type of treater would entirely eliminate the carbon trouble when operating on such gas as was handled at Sault Ste. Marie. Such a deposit was not found in any instance previously.

The following explanation of the phenomenon is offered. Temperatures up to those obtained in city gas plants are not high enough to crack an appreciable portion of the gas, or its suspended contents, into free carbon and gas; though the fog particles may be decomposed into particles of pitch containing a large proportion of free carbon.



It was not determined whether at temperatures attained in a coke oven, the cracking of the tar fog particles into the elements carbon and hydrogen, would occur. It is obvious, however, that fire in an oven or main would induce only incomplete combustion of the tar, and would leave large proportions of soot suspended in the gas. It is doubtful if these particles, of soot, as distinct from tar or pitch, would be found in gas from ovens working normally.

After several unsuccessful attempts to prevent the deposition of carbon, or to devise a means of flushing it off with light oil, it was decided that the insulator must be cleaned periodically by wiping or burning. The insulators used have the usual rough surfaces found on silica combustion tubes. From these pitted surfaces it was impossible to remove the carbon except by burning. At our suggestion the manufacturers undertook to glaze the surface of the quartz insulators. The two tubes, obtained before the work had to close, exhibited pitted surfaces in spots where the glazing had been imperfect. Nevertheless, it was possible to demonstrate that the glazed surface will give satisfaction. From such a glazed quartz surface the carbon can be removed by rubbing with an oiled rag, if this is not delayed until the current has begun to discharge over it. It might also be possible to clean the insulator without opening the chamber, by using a wiper on the end of a rod passing through a gland in the wall. In any case no arduous attendance is required and, in all probability, cleaning would be necessary less than once a shift.

#### *Treater Capacity.*

As pointed out above, the volume of gas that can be cleaned in the treater is limited by the free carbon particles carried, and not by the tar fog. "Pounding" of the exhauster occurs before the treater capacity for tar precipitation has been reached.

The only available method of measuring the gas was by means of a Pitot tube in the pipe leading from the treater to the cooler. No such measurements in small pipes are very accurate; but in this case they were made more difficult by the pulsations of the exhauster. However, it was definitely shown that only a small quantity of free carbon, and practically no tar, would be left in gas that had passed through the pipe at as high a rate as 43 linear feet per second. Disregarding the water vapour content, this implied a capacity of one and one-half million feet of gas at 60° F. per day. As there is no meter in the plant

to measure the total gas production, and no main long enough to give satisfactory Pitot tube readings, it was impracticable to do more than estimate the gas handled by the number of tons of coal used per day. An approximate check, also, was obtained from comparing the total tar produced with that collected in the treater. Assuming that 60 per cent. of the total tar comes down in the collecting main, the rate of tar collection in the treater indicated that it, the treater, would satisfactorily clean the gas produced from between 100 and 125 tons of coal per day. Taking 1,500,000 cubic feet as the total gas volume, this means a production of between 12,500 and 15,000 cubic feet of gas per ton of coal in the ovens. Average gas analyses indicate that this is approximately correct. It is not to be recommended that any installation should be made in which each pipe is required to handle more than a million feet per day.

Filtration measurements were made to determine the percentage efficiency of the cleaning, as compared with that given by the tar extractors of the plant; taking two milligrammes of carbon per cubic foot of gas as a satisfactory maximum, it was found that gas from the treater contained from one to three milligrammes per cubic foot, whilst gas from the outlet of the tar extractors showed from eight to nineteen milligrammes per cubic foot.

#### *Character of By-products Collected.*

The liquor collected in the treater consisted of tar at approximately 25 per cent water. The amount of water thus collected could be reduced, or possibly eliminated, by insulating the treater pipe, as the gas was well above the temperature at which the water vapour present would saturate it. After settling, the tar contained about  $7\frac{1}{2}$  per cent of water.

The light-oil content of the tar collected in the treater was identical with that collected in the plant. Hence it appears, as was expected, that tar fog particles will not absorb appreciable quantities of light oil in cooling from 85°C. to 30°C. Presumably this statement does not apply to the smaller quantities of benzol present; this, however, would be saved in the benzol recovery plant.

The ammonia liquor collected in the cooler handling the treater gas was very much lighter than that produced ordinarily in the plant. The latter liquor contained an appreciable quantity of finely divided tar, that would settle if the liquor were undisturbed for any length of

time, but which would remain in suspension with the slightest agitation. This suspended tar becomes a serious nuisance in the operation of the ammonia stills. *The ammonia liquor collected from the treated gas was entirely free from particles of suspended tar.*

No conclusions were reached as to the effect of electrical precipitation without cooling, on the problem of naphthalene deposits. Interesting speculations on this question are given in the paper by Professor White. Accurate data can be obtained only by a continuous run on the total gas produced in the plant.

#### CONCLUSION.

Successful tests of the process of electrical precipitation for the de-tarring of gas on a commercial scale have thus been made in wood, coal, and petroleum distillation plants. Various advantages peculiar to this process are mentioned in the course of this paper. The outstanding fact is that installation and renewal costs of by-product collection apparatus may be much reduced, the by-products themselves can be collected in much more desirable condition for refining, and operating costs not affected by the use of electrical precipitation. The results of the tests are so satisfactory, both technically and financially, that it is highly desirable that installations of one of each of the various types of distillation plants be made at once. It is to be urged that the work be not undertaken simply to provide an auxiliary to present systems; but essentially for the purpose of developing a more efficient by-product collection process.

Various expressions of appreciation for assistance given will be found in the course of the paper. In addition, acknowledgement is herewith made of the co-operation of the Western Precipitation Company in the form of valuable advice and occasional financial assistance. The writer also wishes to acknowledge his indebtedness to Mr. J. D. Chase, who has given him invaluable assistance during the course of the work subsequent to that done in the Vancouver gas plant.

UNIVERSITY OF BRITISH COLUMBIA,

June 22, 1918.

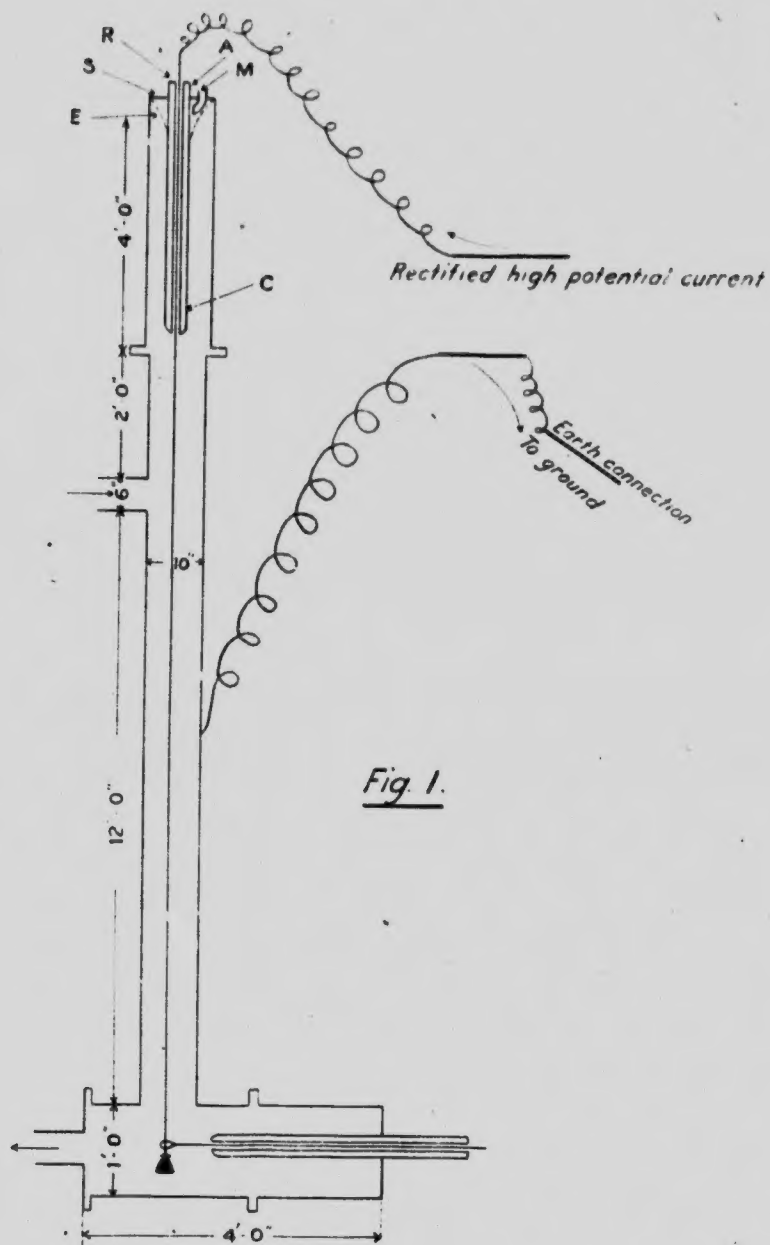


Fig. 1.

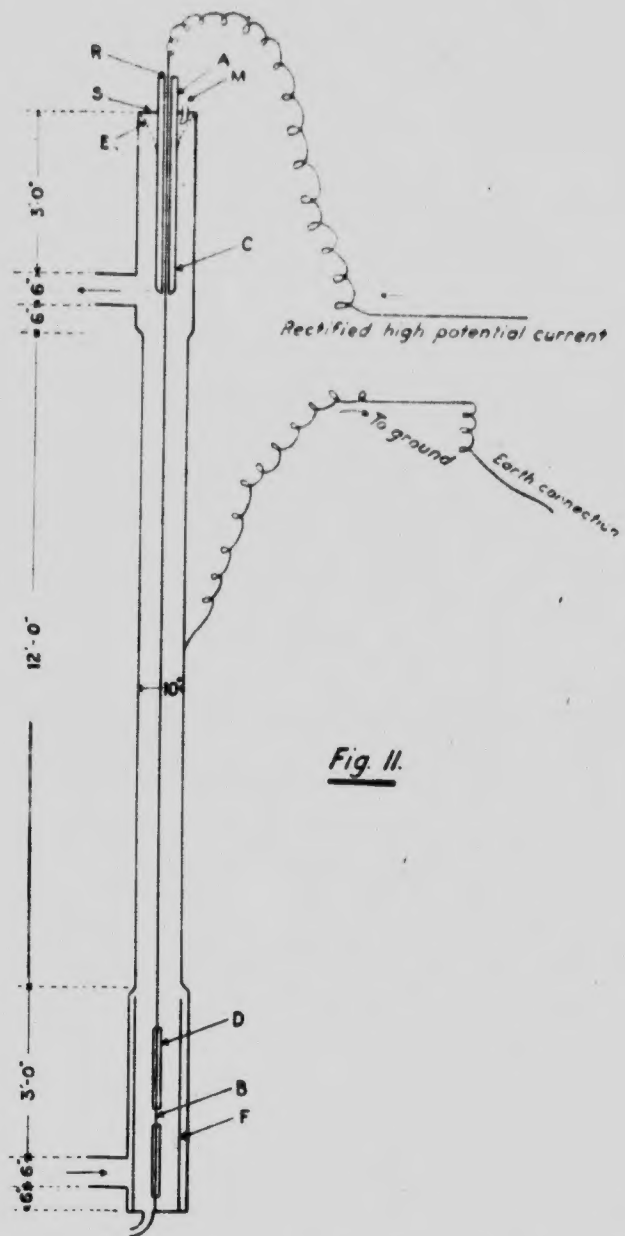


Fig. 11.

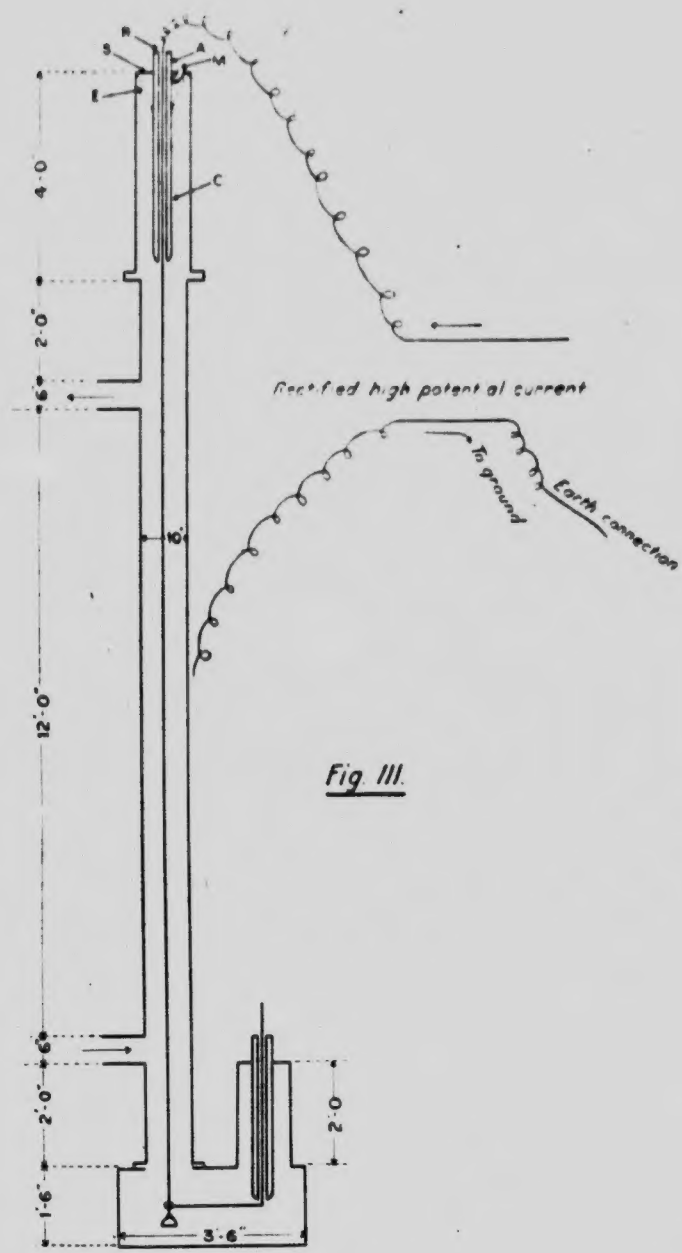


Fig. III.

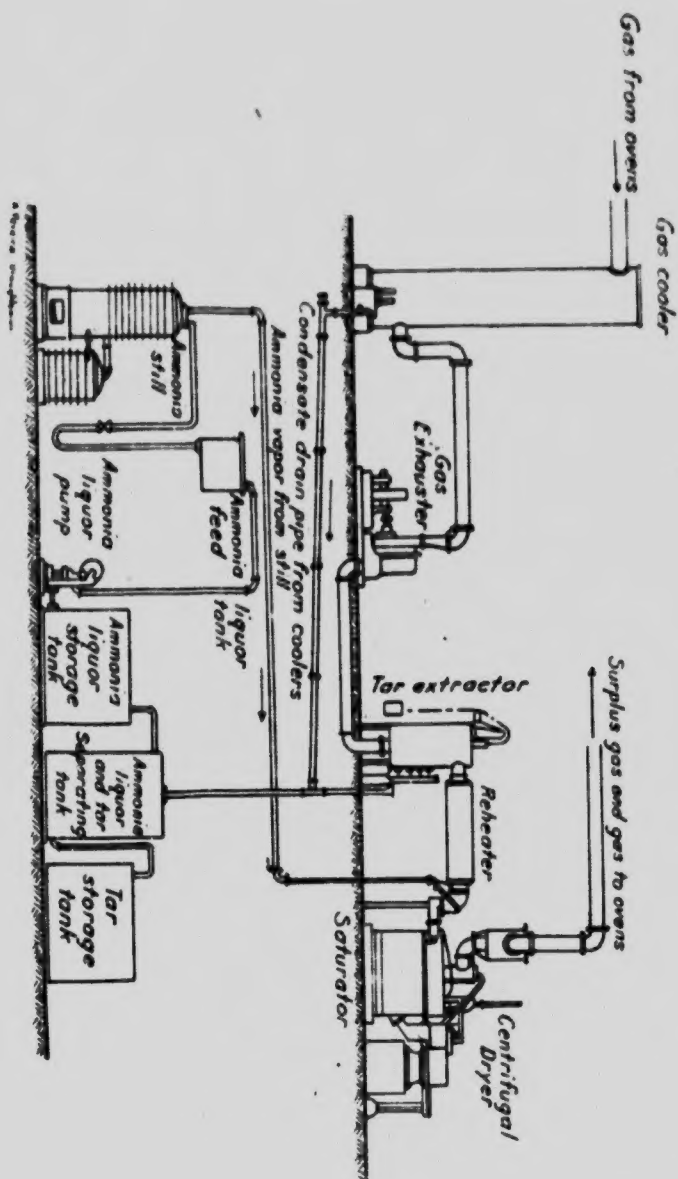


Fig. IV. Koppers Patent.